1.0 OAK RIDGE GASEOUS DIFFUSION PLANT MASS BALANCE PROJECT

1.1 PROJECT OVERVIEW

This report has been prepared to summarize the findings of the Oak Ridge Gaseous Diffusion Plant (ORGDP)[1] Mass Balance Project and to support preparation of associated U. S. Department of Energy (DOE) site reports. The project was conducted to support DOE efforts to assess the potential for health and environmental issues resulting from the presence of transuranic (TRU) elements and fission products in recycled uranium (RU) from reactor returns that was processed by DOE and its predecessor agencies. The U.S. Government used uranium in fission reactors to produce plutonium and tritium for nuclear weapons production. Because uranium was scarce relative to demand when these operations began almost 50 years ago, the spent fuel from U.S. fission reactors was processed to recover uranium for recycling.

Uranium that has been irradiated in reactors contains TRU elements [e.g., plutonium (Pu) and neptunium (Np)] and fission products [e.g., technetium-99 (^{99}Tc)]. Following chemical processing to recover uranium for reuse, trace quantities of Pu, Np, and ^{99}Tc continue to be present in the RU stream. These contaminants make the RU stream more radioactive than natural uranium. The processing and re-enrichment of RU thus may potentially introduce health and environmental consequences beyond those associated with the uranium stream.

In response to these concerns, DOE initiated an effort to identify all situations in which the processing of RU by DOE and its predecessor agencies could have created exposure hazards for workers and/or significant contamination to the environment. The first step in this process involves the "mass balance review." The mass balance review attempts to determine how much RU was generated by the U.S. Government during a period of approximately 47 years and to determine how it was distributed, processed, and used. DOE's reconstruction of the historical flow and processing of RU has three fundamental elements:

- Determining annual mass flow of RU throughout the DOE system from the start of processing to March 31, 1999.
- Identifying the characteristics and contaminants (e.g., Pu, Np, and ^{99}Tc) in the major uranium streams.
- Conducting at appropriate sites mass balance activities sufficient to identify any significant implications for personnel exposure or environmental contamination.

The DOE mass balance review includes U.S. Government sites that were sources for RU (i.e., that processed irradiated fuel to recover uranium for recycling); sites that re-enriched the RU stream in the fissile ^{235}U isotope; sites that manufactured weapons components; and other affected sites. As the first U.S. gaseous diffusion plant (GDP) for enriching uranium in the ^{235}U isotope for weapons and for commercial nuclear fuel, ORGDP had significant involvement

[1] Following the shutdown of ORGDP in 1987, the facility was known as the Oak Ridge K-25 Site. In 1997, it became the East Tennessee Technology Park (ETTP).

with the flow of RU materials from and to other sites. ORGDP's involvement with other sites included:

- Receiving RU from U.S. Government facilities at Hanford and Savannah River and from Harshaw Chemical Company, following use of chemical separation processes to extract uranium from irradiated fuel.
- Receiving RU from other U.S. GDPs—the Paducah Gaseous Diffusion Plant (PGDP), which shipped RU material as UF_6 feed to ORGDP, and the Portsmouth Gaseous Diffusion Plant (PORTS), which provided much smaller quantities of RU as UF_6 feed.
- Receiving RU from commercial enrichment customers (primarily nuclear utilities in France, the United Kingdom, and Germany).
- Receiving partially enriched UF_6 product from PGDP that contained certain RU constituents (although this PGDP-enriched product was technically not RU).
- Converting RU received to uranium tetrafluoride (UF_4) and to uranium hexafluoride (UF_6) for use as GDP feed.
- Shipping RU converted to UF_6 or UF_4 to PGDP and PORTS.
- Shipping RU fluorination tower waste ashes from the UF_6 conversion process to PGDP (which subsequently shipped them to Fernald).
- Shipping product enriched in the ORGDP cascade to the Y-12 Plant, PORTS, and to private-sector companies fabricating fuel for commercial enrichment customers.
- Shipping tails from the ORGDP enrichment cascade to PGDP for additional "stripping" in the PGDP cascade.
- Shipping RU from commercial enrichment customers to PGDP for storage after ORGDP was placed on standby (without re-enriching the RU in the ORGDP enrichment cascade).
- Shipping cylinder heels to PGDP after ORGDP was placed on standby.

ORGDP began gaseous diffusion enrichment production in 1945. The subsequent processing of RU, which began in 1952, impacted a broad range of processes and activities in facilities and locations across the ORGDP site. Facilities with significant involvement in RU processing included UF_6 feed production facilities; a waste ash pulverization and uranium recovery facility; decontamination and uranium recovery facilities; facilities fed by waste streams from decontamination facilities; other facilities performing more limited uranium recovery and decontamination activities; and the ORGDP enrichment cascade, which operated in a variety of different configurations over time.

In 1985, with an overcapacity of enrichment capabilities, ORGDP was placed on standby status. The plant was officially shut down in 1987. Recently, DOE has initiated a program of reindustrialization at the site, which in 1997 became known as the East Tennessee Technology Park (ETTP). ETTP also serves as the base of operations for environmental activities at facilities managed by DOE Oak Ridge Operations (DOE-ORO).

The ORGDP Mass Balance Project represents an effort to collect, verify, analyze, and interpret available data to provide an overall accountability, or site mass balance, for ORGDP RU streams. In addition, data on ORGDP processes and activities and data on Pu, Np, and ^{99}Tc—the primary contaminants of concern in the RU stream—have also been collected, analyzed, and interpreted. Based on this information, the project team has attempted to identify all those activities that (1) created a likelihood of workers coming into contact with concentrated

RU constituents through direct physical contact or via airborne dust and/or (2) caused reportable environmental releases of concentrated RU constituents.

1.2 PURPOSE AND SCOPE

The purpose of the ORGDP Mass Balance Project is to support DOE's efforts to identify all situations in which the U.S. Government processing of RU could have created exposure hazards for workers and/or significant contamination to the environment. Following guidance provided in DOE's *Historical Generation and Flow of Recycled Uranium in the DOE Complex: Project Plan*, the ORGDP project team has focused on:

- Describing the amounts, characteristics, and constituents of the incoming and outgoing RU streams at ORGDP.
- Understanding the historical processes, product specifications, and process activities that concentrated the primary RU constituents of concern (Pu, Np, and ^{99}Tc).
- Determining the facilities and processes that could cause worker exposures or lead to measurable environmental contamination.
- Performing annual mass balances for RU and for Pu, Np, and ^{99}Tc to the degree existing mass and analytical data permit.

The project has identified and reviewed RU streams at ORGDP from the initial introduction of RU into the plant until March 31, 1999. At ORGDP, these streams encompassed a broad spectrum of material forms, including uranium dioxide (UO_2), uranium trioxide (UO_3), UF_6, UF_4, feed conversion ash, scrap, and a wide variety of other associated wastes. The RU flow has been traced from receipt by ORGDP until ultimate disposition by the plant. Efforts have also been made to identify all other DOE sites with which ORGDP interfaced via RU streams and to determine how the plant worked with them.

To place emphasis on the RU flows that most warrant attention, the project team has followed the guidance of the DOE *Project Plan* and identified the RU flows that posed no significantly increased hazard. These RU streams contained Pu, Np, and ^{99}Tc constituents that would have provided an incremental radiological dose of significantly less than 10% of the dose provided by the uranium itself. These streams also represented final product or waste forms, with no additional processing anticipated. DOE has deemed such end products to be outside the scope of the mass balance project. The process for identifying these RU streams, which include ORGDP enrichment cascade product and tails streams, is documented in this report.

1.3 PROJECT IMPLEMENTATION STRATEGY

An interdisciplinary project team was formed to conduct the ORDGP Mass Balance Project. Team members included individuals with extensive experience in DOE uranium enrichment operations; uranium processing; nuclear materials control and accountability; health and safety at DOE facilities; nuclear engineering; the nuclear fuel cycle; statistical analysis; and data and information management. Guided by information provided in the DOE *Project Plan* (e.g., the Question Set and the Site Report Outline), the team developed a strategy and process for identifying, collecting, organizing, and analyzing available data relevant to the project. Leads were established for major project areas (e.g., site historical overview, RU mass flow, mass balance activities, and constituents in RU), and team members were designated to research and abstract information on specific topics. Formal team meetings were held twice each week to track progress and discuss project issues.

The project team searched a variety of data collections and libraries at ETTP and other Oak Ridge Complex locations to identify and retrieve data. Major data sources consulted and analyzed included:

- Nuclear Materials Control and Accountability (NMC&A) Material Balance Reports, including shipping, receiving, and inventory records.
- Nuclear Materials Management and Safeguards System (NMMSS) data.
- ORGDP historical site reports, including quarterly plant reports and engineering progress reports.
- ORGDP reports describing facilities and production processes.
- Plant records, including employment and health physics records.
- ORGDP production records.
- ORGDP analytical laboratory records.
- Correspondence between shippers and receivers.
- Historical DOE and contractor reports addressing RU.
- More recent (i.e., post-1990) health physics reports on the site.
- More recent environmental survey reports on the site.
- Interviews with ORGDP personnel or with personnel with direct experience with enrichment operations.

Where NMC&A data was unavailable, attempts were made to obtain NMMSS data to fill the gaps. Team members worked with representatives of other DOE sites with which ORGDP interfaced via RU streams to verify shipping and receiving data and reconcile differences between sites. Any unresolved data discrepancies will be turned over to the DOE Working Group Team for assistance with resolution.

In addition to consulting the ORGDP analytical laboratory records, the team found it necessary to glean analytical data from a wide variety of sources, including the ORGDP historical quarterly reports and health physics reports. Correspondence between shippers and receivers also provided a record of comparisons of sets of analytical data, the first set developed by the site shipping RU and the second by the site receiving the material. In addition, analytical data has been compared and shared with other appropriate DOE sites.

For some areas that presented gaps in data that could not at present be filled by research, the project team developed estimates for quantities of RU and/or constituents. These estimates are based on extrapolations from actual data and represent (1) application of known data from similar material and/or circumstances or (2) application of known data from a specific time period over a longer or a shorter period of time. All such estimates and their bases are specifically identified in this report.

The project team analyzed data on receipts, shipments, inventories, product, tails, releases, and other categories—along with available analytical data—in the context of documented historical information on ORGDP processes and activities. Understanding of GDP processes known to concentrate Pu, Np, and ^{99}Tc and of GDP processes and activities known to create potential for exposure to these RU constituents provided additional context for analysis. By correlating mass balance data, analytical data, health physics data, environmental sampling data, and historical information on ORGDP processes, the team was able to identify specific processes, locations, and time periods of importance for potential worker exposure or environmental contamination. These processes, locations, and time periods became the focus of additional assessment to determine the situations that had the potential to create exposure hazards for workers and/or significant environmental contamination.

The ORGDP project team has made an effort to identify and resolve discrepancies between the findings presented in this report and information found in other DOE site reports and previous DOE historical reports. This report attempts to identify major discrepancies and describe their resolutions.